Comment on arXiv:1105.6334 "Optical nonlinearity in Ar and N_2 near the ionization threshold"

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In a recent publication [Phys. Rev. Lett. **107**, 103901 (2011)], Wahlstrand *et al.* reported to observe no indications for the appearance of the higher-order Kerr effect in a parameter regime that was previously found to display this phenomenon [Opt. Express **17**, 13429 (2009)]. Here we show that careful analysis of the original experimental data of Wahlstrand *et al.* reveals a 22% saturation, i.e., direct proof for the appearance of the higher-order Kerr effect. In the light of these findings, the validity of Wahlstrand *et al.*'s main conclusions appears highly questionable.

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In a recent publication [1], Wahlstrand *et al.* revisit the appearance of the higher-order Kerr effect (HOKE) in argon and nitrogen at intensity levels up to 180 TW/cm^2. Using spectral interferometry, they monitor plasma-induced as well as instantaneous contributions to the nonlinear refractive index. In contrast to previous affirmative reports [2], however, Wahlstrand *et al.* see no saturation due to the HOKE in their measured data. Here we show that their original data contains ample evidence of Kerr saturation, which was overlooked in their previous analysis.

Directly using the vector data embedded in Figs. 2(c) and (d) of [1], we retrieved the measured refractive index change Δn as a function of temporal delay Δt between a pump and a probe pulse for intensities of 15, 120, and 180 TW/cm^2, both for parallel and for perpendicular polarization, see ancillary files linked to this submission. As there is no plasma formation at the lowest intensity of 15 TW/cm^2, the corresponding curves exhibit an isolated instantaneous Kerr response, which we exploit for direct reconstruction of the temporal pulse shape without the need for any model assumption or approximation. At higher intensities, plasma contributions to the nonlinear phase appear. For these cases, we computed the index changes due to plasma formation using PPT theory [3]. Our calculations indicate a serious discrepancy in the intensity calibration in Fig. 2 of Ref. [1], with intensities being 1.5 to 2 times higher than compatible with PPT theory. The analysis was repeated for several other ionization models, including ADK and simple multiphoton laws, which, however, leaves the following conclusions unaffected.

Using the same approach as utilized by Wahlstrand *et al.* in their Fig. 3(c), we separated plasma and Kerr response [see our Fig. 1(a) and (b) and ancillary data files linked to this submission]. The peak phase shifts of the isolated Kerr response are compiled in Fig. 1(c). For both polarizations, the Kerr contribution to the nonlinear phase clearly saturates above 120 TW/cm^2, with the phase shifts at 180 TW/cm^2 lying 22±5% below a linear extrapolation based on measured data at lower intensities [dashed lines in Fig. 1(c)]. The error margins and the significance of this saturation effect were checked by repetition of the analysis with different ionization models. At 180 TW/cm^2 and for parallel polarization, this amounts to a HOKE contribution of -50 mrad at zero delay, i.e., the HOKE is about half as strong as the negative index contribution from the generated plasma. It is important to recall that our analysis of the data in [1] is based on their measured data only, is extremely robust against choice of the ionization model, and is free of any other model assumptions.

Our analysis therefore refutes the main conclusion of Wahlstrand *et al.* on the irrelevance of the HOKE in nonlinear optics and filamentation. In fact, it seems that Wahlstrand *et al.* delivered further experimental evidence for the Kerr saturation, even though apparently at higher intensities than previously observed. We believe that differences in the experimental approach may contribute to this disagreement, which strongly suggests a need for further and more thorough experimental investigations of the HOKE.

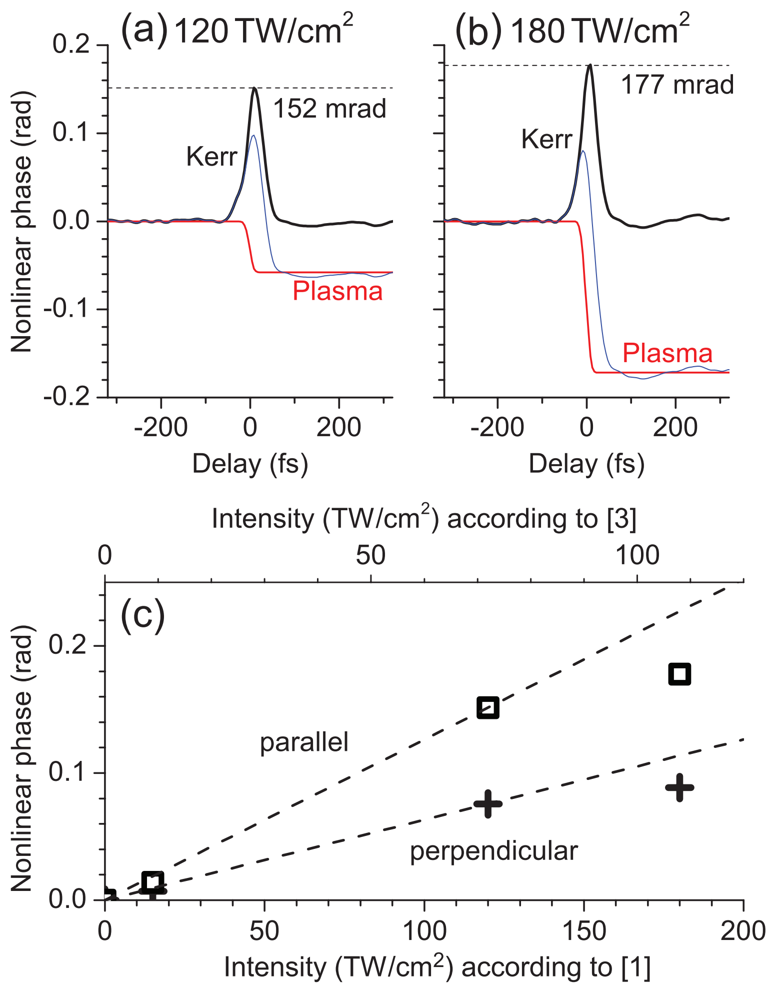

FIG. 1: Kerr saturation in the experimental data of [1]. (a) Separation of Kerr response (thick black line) and plasma response (thin red line) for the 120 TW/cm^2 trace (thin blue line) in Fig. 2(c). (b) same for 180 TW/cm^2. Dashed lines indicate peak Kerr phase shift. (c) Compilation of all retrieved peak phase shifts (squares: parallel pump-probe polarization, crosses: perpendicular polarization). Linear extrapolations from the low power data are shown as dashed lines. Bottom axis refers to intensity calibration of [1], top axis to PPT-compatible units.

[1] J. K. Wahlstrand, Y.-H. Cheng, Y.-H. Chen, and H. M. Milchberg, `arXiv:1105.6334v3 [physics.optics]` and Phys. Rev. Lett. **107**, 103901 (2011).

[2] V. Loriot, E. Hertz, O. Faucher, and B. Lavorel, Opt. Express **17**, 13429 (2009).

[3] A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, Sov. Phys. JETP **23**, 924 (1966).